Defining source-receptor relationships for mercury: measurement and modeling approaches

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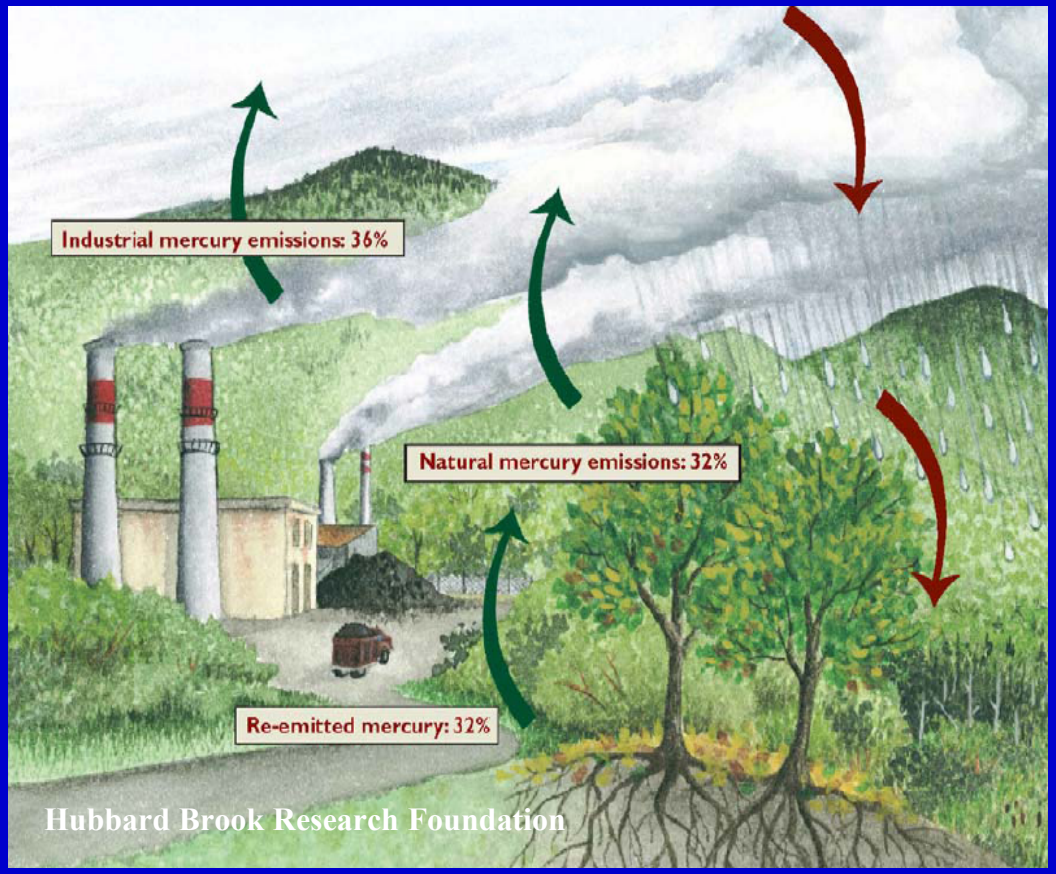
Emissions, Transport and Deposition

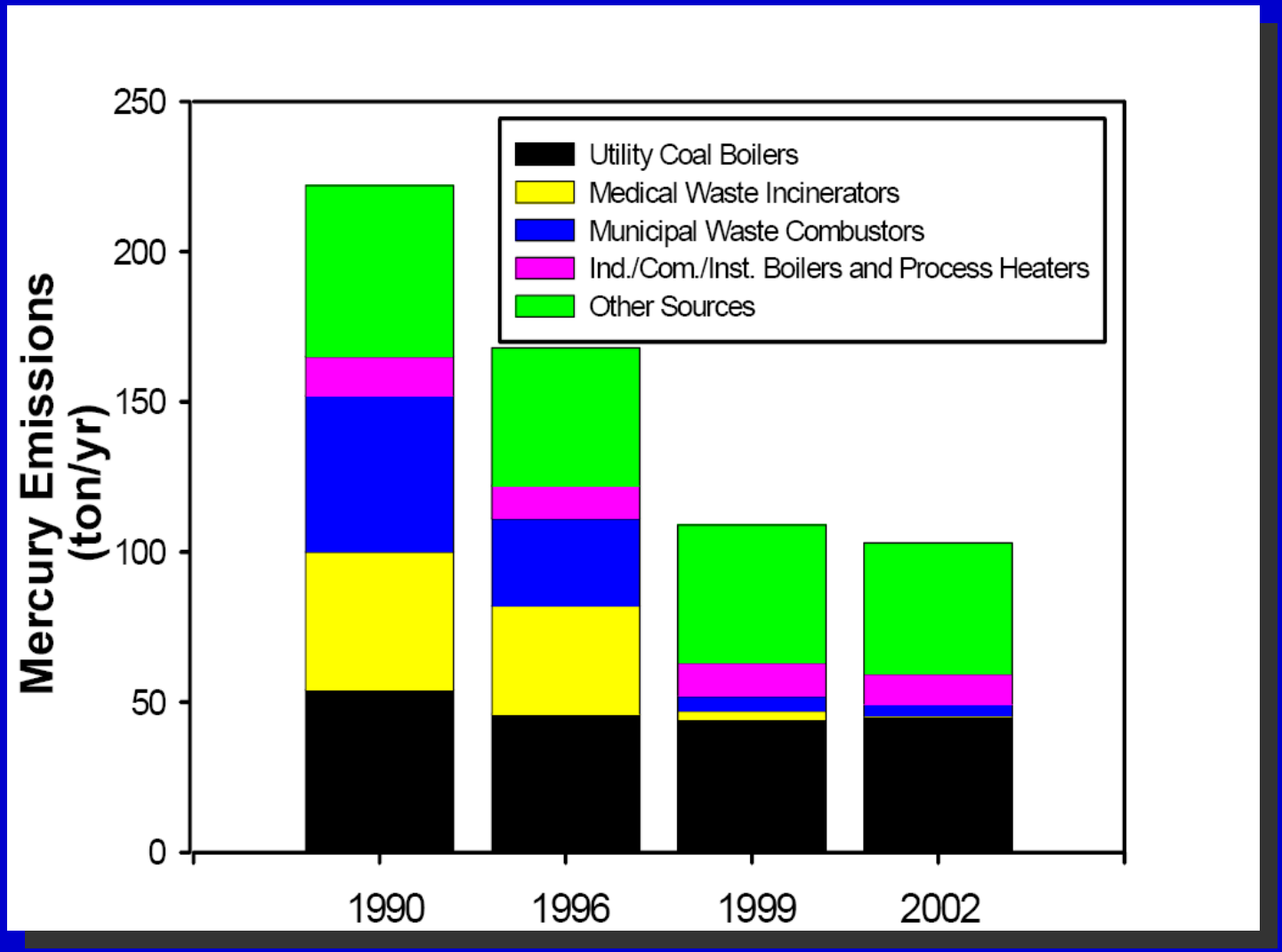


Key Questions

- Where does the mercury in U.S. fish and wildlife mostly originate from?
- Is it U.S. sources or global sources?
- Is this true for marine as well as freshwater fish?

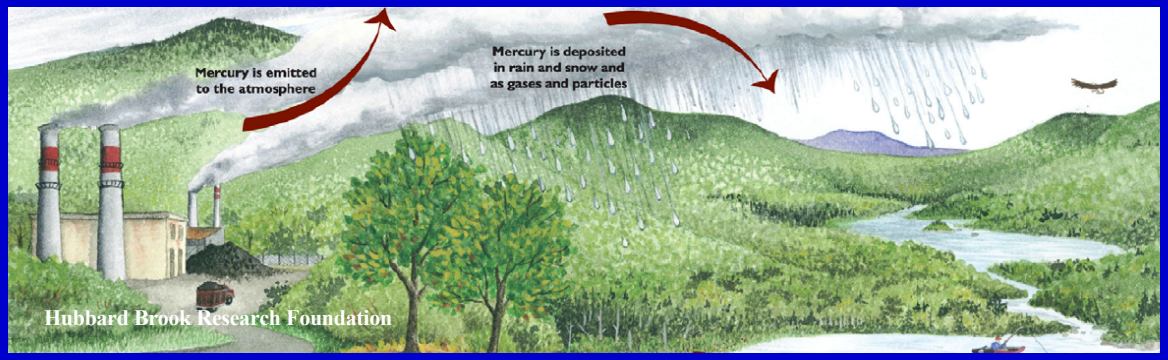
Speciation of the mercury controls it's fate.

Mercury Emissions in U.S.



Source Apportionment

-relates sources and environmental concentrations.



Approaches

Source modeling (e.g., CMAQ) – source to receptor

- Requires emission inventory, chemistry, and meteorology
- Models emission source impacts on predicted concentrations

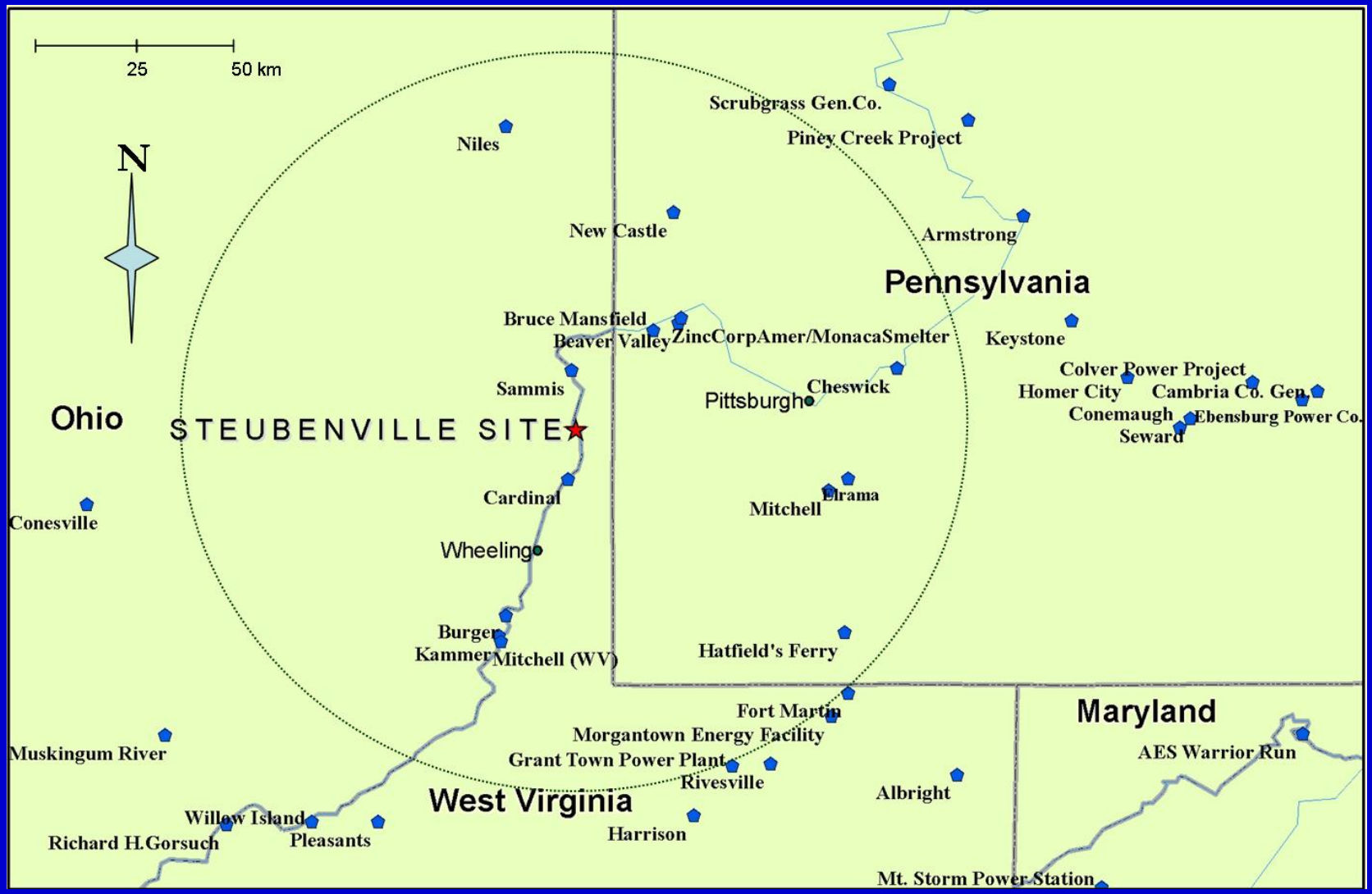
Receptor modeling (e.g., PMF) – receptor to source

- Requires comprehensive environmental measurements.
- Statistically identifies sources impacting measured concentrations.
- Includes meteorological information including NEXRAD.

Steubenville Mercury Study

- **Objective**
  - Determine the impact of local/regional coal combustion sources on mercury deposition in the Ohio River Valley.
- **Mercury Study Milestones**
  - Study designed in 1999.
  - Research funded under competitive cooperative agreement with EPA ORD.
  - 4-years of data collection.
  - 2-years of wet deposition data analysis and modeling completed (2003-2004).
    - Keeler *et al.,* 2006 ES&T **40**, 5874-5881.

Location of Surrounding CFUBs



Study Approach

Collected detailed measurements

- Speciated Ambient Mercury-continuous
- Event-based wet deposition sampling
- Potential source co-pollutants (trace elements)
- On-site Meteorology
- Aerosols - Integrated and Continuous
- Criteria Gases – Continuous

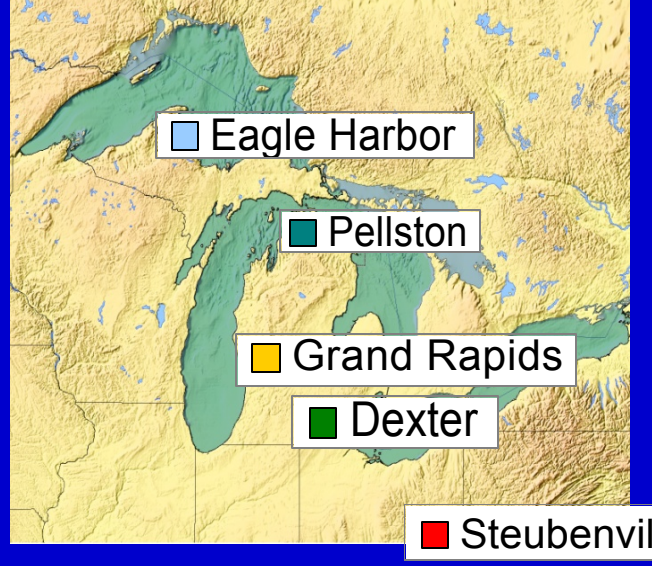
Applied state-of-the-art receptor models

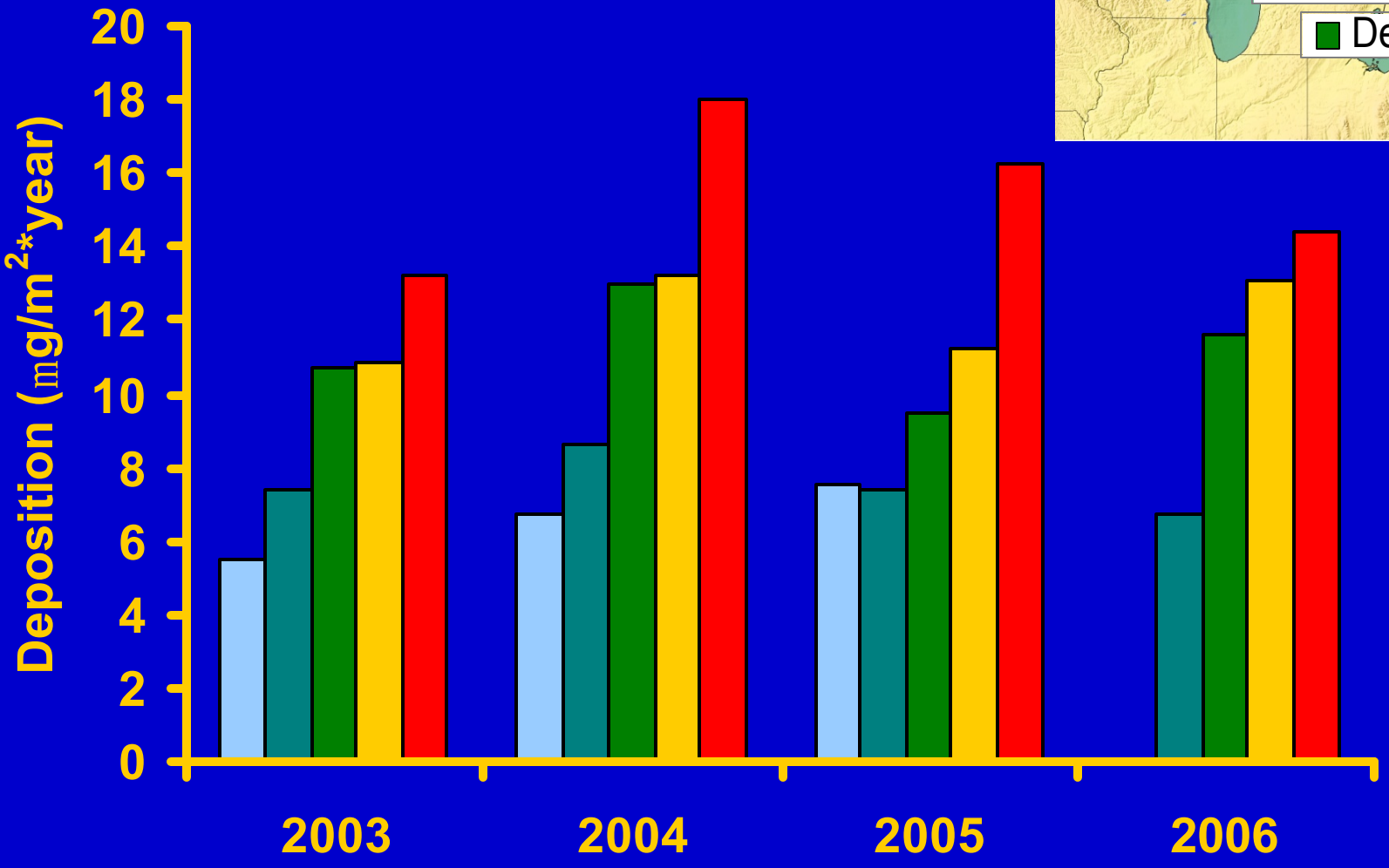
- Mercury source apportionment demonstration
- Latest version of EPA models – UNMIX & PMF
- Hybrid Modeling (Regional Transport)
- Detailed Storm Analysis -NEXRAD

Summary of Steubenville Results

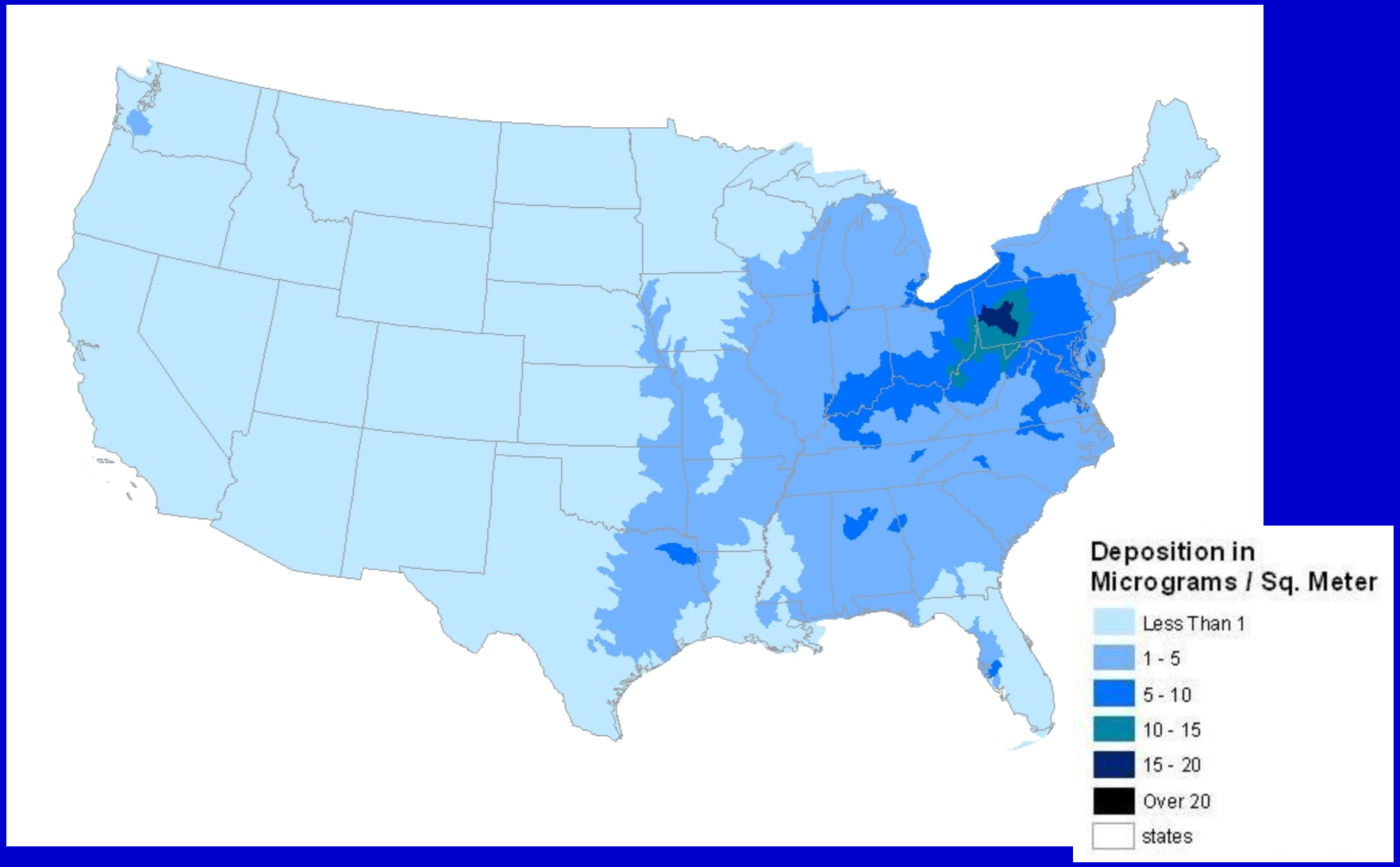
- **Mercury wet deposition at Steubenville**
  - ~ 80% is attributable to local/regional anthropogenic sources
  - ~ 70% is attributable to coal combustion
  - ~ 20% from re-emission or global background
- **A significant fraction of the Hg wet deposition is driven by a few local coal combustion dominated precipitation events;**
- **Rapid removal of RGM observed at onset of rain;**
- **Dry deposition even more local in origin.**

2003 – 2006 Great Lakes Deposition Comparison





Mercury Deposition From US Power Plants in 2001



Source: US EPA 2005 using Community Multiscale Air Quality Model.

Comparison of USEPA CMAQ Results and Measured Mercury Wet Deposition at Steubenville

	Hg Deposition ($\mu g\ m^{-2} y^{-1}$)	CFUB? Contribution (%)
CMAQ 2001	13.6 (modeled)	43
PMF/UNMIX 2003-2004	16.5 (measured)	72

?CFUB-Coal-fired Utility Boiler

CMAQ Simulations performed by CSC for EPA (6FEB04)

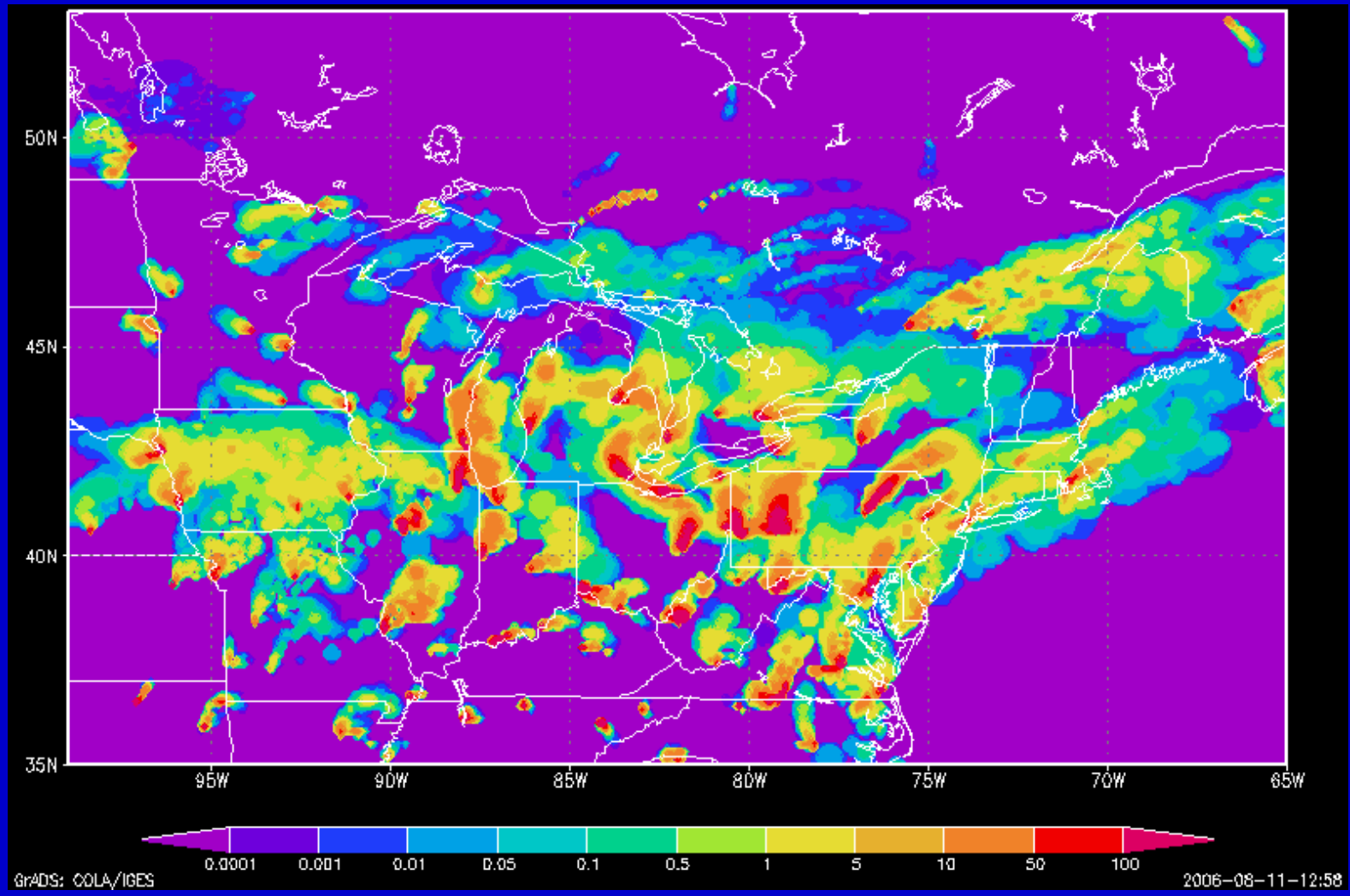
CMAQ Modeled vs Measured Event Hg Wet Deposition
in 2001

Site	CMAQ Wet Deposition ($\mu g\ m^{-2}$)	Measured ($\mu g\ m^{-2}$)
Dexter, MI	8.3	12.5
Pellston, MI	4.6	10.5
Eagle Harbor, MI	4.7	7.7
Underhill, VT	4.4	8.6

CMAQ Results provided by Russ Bullock., USEPA

Dry Deposition of Hg

August 10-11, 2006



Significance of Results

1. Current models (including those used by EPA for CAMR analyses) estimate a much lower local/regional source contribution to Hg deposition, on average:
   - About 8% of domestic Hg deposition estimated to be from domestic electric utility coal combustion.

2. Implications for potentially vulnerable areas (i.e., "Deposition Hotspots"), which will not be identified by current national network.

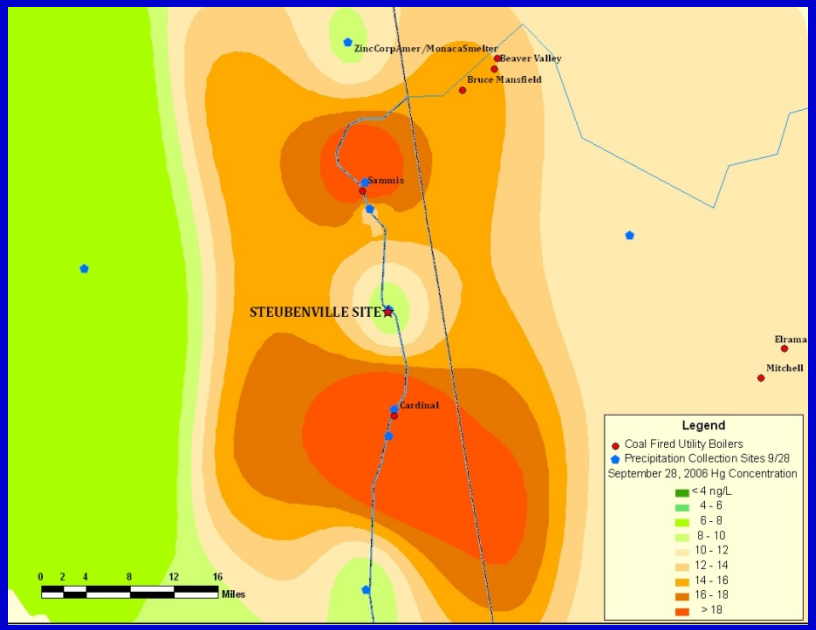
3. Significant deposition decreases predicted for Steubenville area.

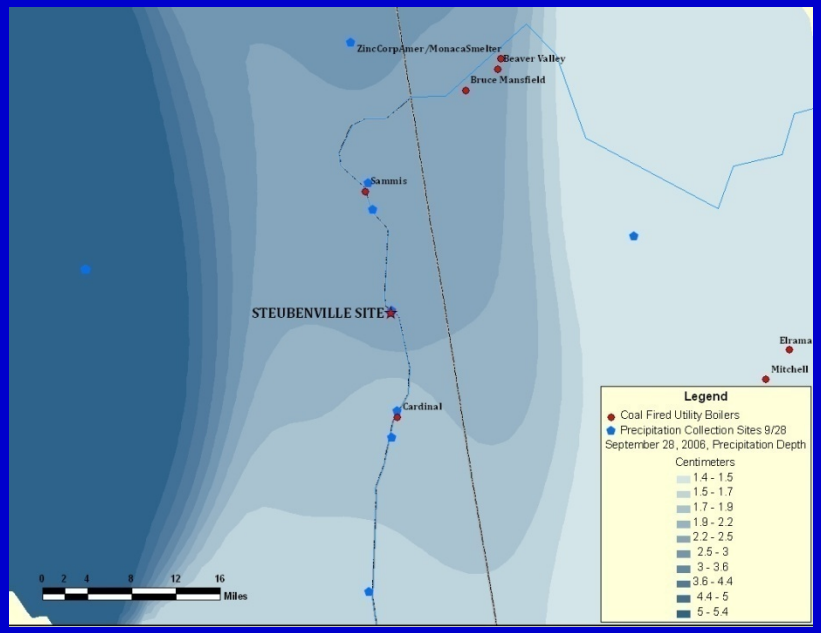
Why are actual deposition values higher than those predicted by air quality models?

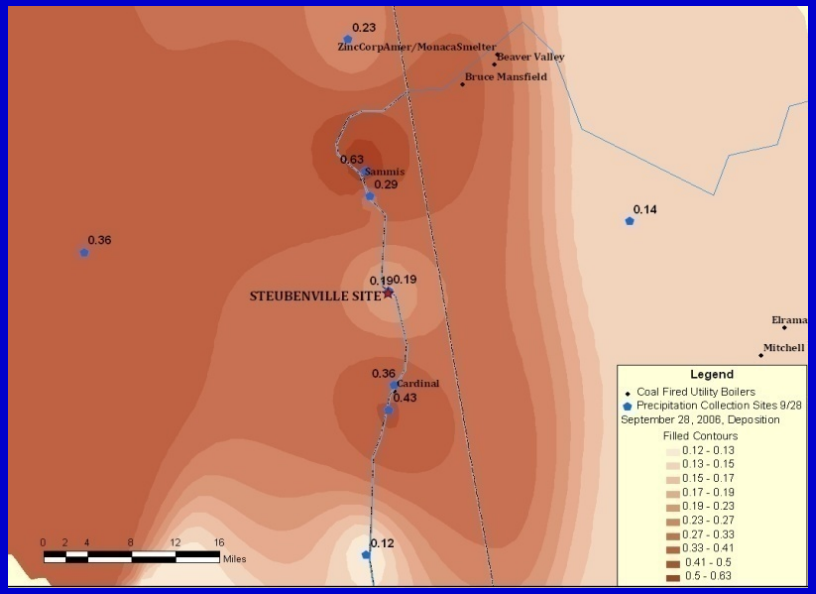
1. Speciated mercury emissions data for major sources still lacking, time resolution annual;
2. The deposition parameterizations in current models are inadequate:
   - High Hg concentrations and deposition in urban areas (e.g., Chicago, Charlotte, St. Louis, and Detroit);
   - Underestimates in predicted deposition; Hg^0 dry deposition poorly described;
   - Hgp size distribution not properly described;
   - Photochemistry not adequately included;
3. Event-based empirical deposition data is lacking, especially on proper spatial scales.

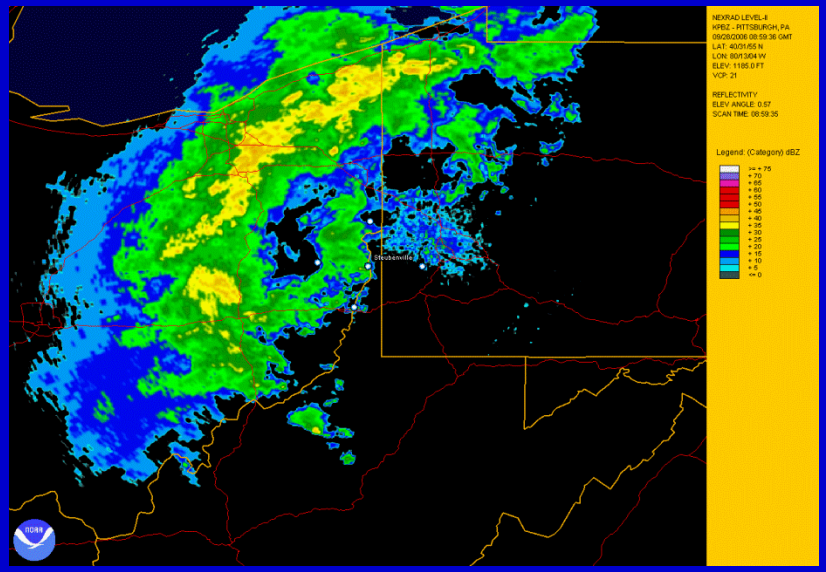
Location of Steubenville Intensive Sites and CFUB

Case Study: September 28









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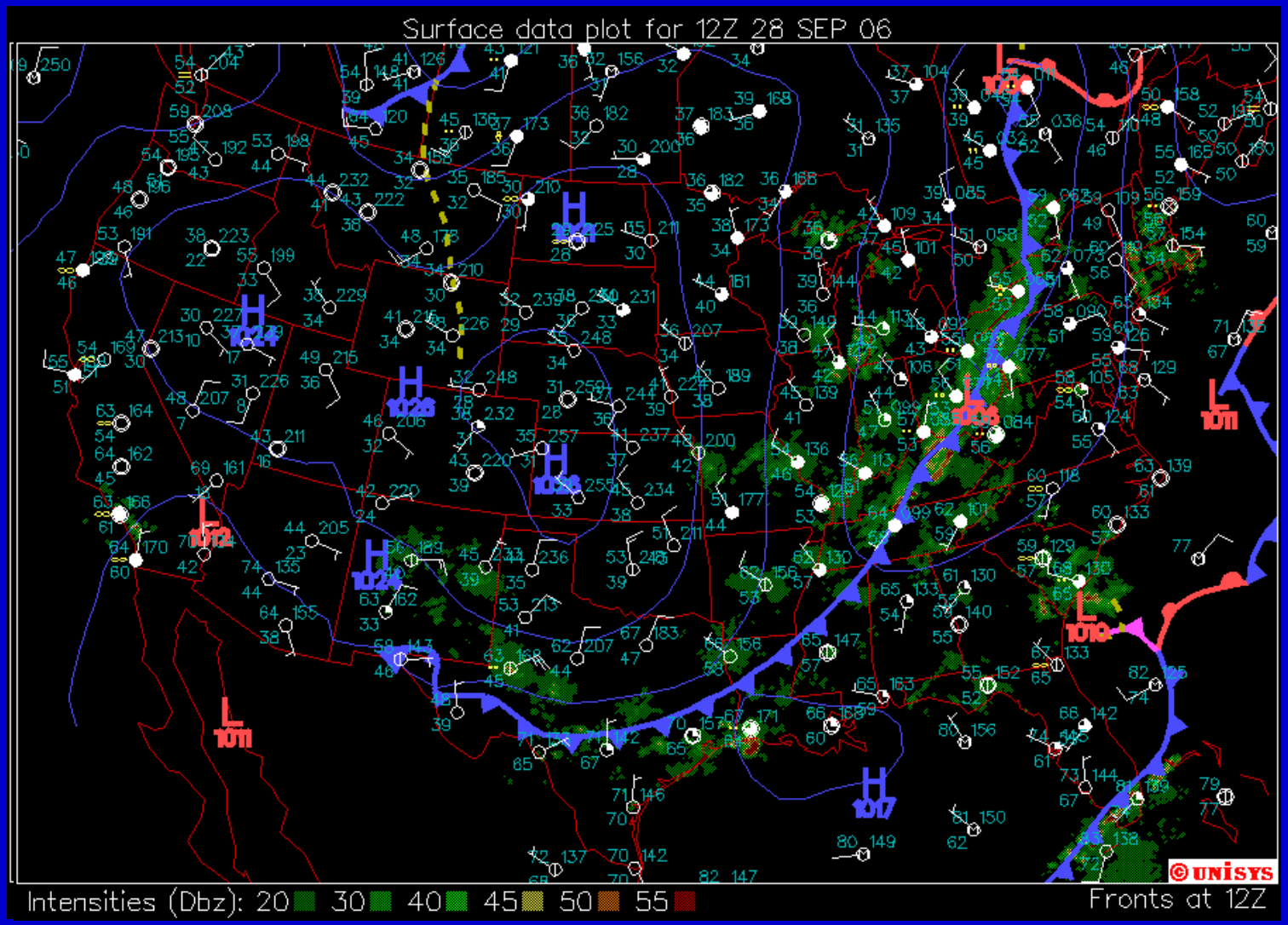
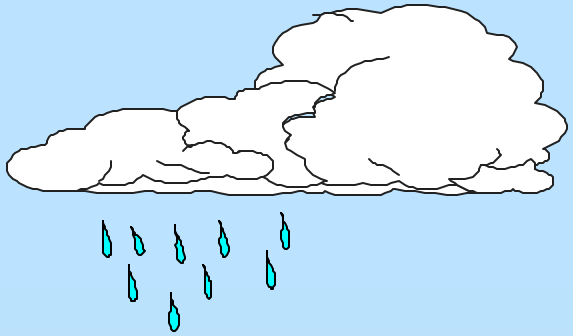
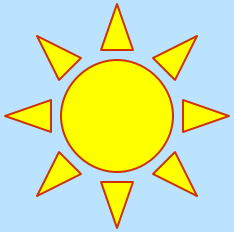


ATMOSPHERIC MERCURY IN THE COASTAL ENVIRONMENT





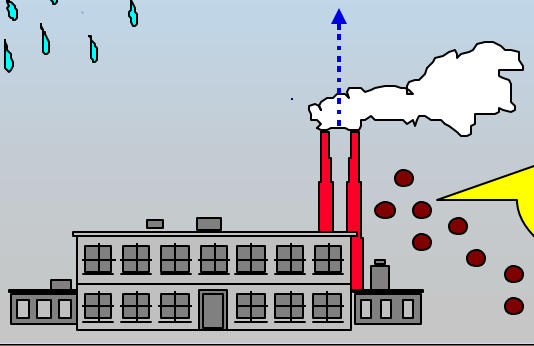
Photochemistry

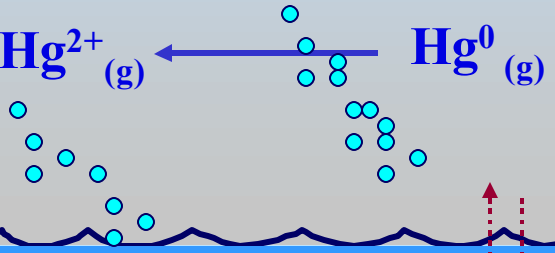
Wet Deposition

$Hg_{(p)}$

Chemical Reactions

$Hg^{2+}_{(g)}$ ← $Hg^{0}_{(g)}$





Sea Spray

Anthropogenic Emissions

Fish Advisories

Summary Points

1. Anthropogenic inputs of mercury to the atmosphere exceed those from natural sources. Its fate after emissions depends on the form of mercury emitted, e.g. RGM or Hg^0

2. Although US emissions are a small fraction of total global emissions, they make a significant contribution to US deposition.

3. Large emission sources can produce areas of high mercury deposition that are not predicted in current national scale models, and are not observed in the national networks.

4. Atmospheric mercury chemistry in coastal regions could enhance mercury chemistry to near-shore environments.

4. Mercury deposition is only part of the story in determining mercury exposure and risk – what happens in the watershed is important to concentrations in fish and wildlife.